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Comparative Plutonium-239 Dose Assessment for Three Desert Sites: Maralinga, Australia; Palomares, Spain; and the Nevada Test Site, USA–Before and After Remedial Action

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COMPARATIVE PLUTONIUM-239 DOSE ASSESSMENT FOR THREE DESERT SITES:
Maralinga, Australia; Palomares, Spain; and the Nevada Test Site, USA—Before and After Remedial Action

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1. Introduction

As a result of nuclear weapons testing and accidents, plutonium has been distributed into the environment. The areas close to the sites of these tests and accidental dispersions contain plutonium deposition of such a magnitude that health authorities and responsible officials have mandated that the contaminated areas be protected, generally through isolation or removal of the contaminated areas. In recent years remedial actions have taken place at all these sites. For reasons not entirely clear, the public perceives radiation exposure risk to be much greater than the evidence would suggest [1]. This perception seems to be particularly true for plutonium, which has often been "demonised" in various publications as the "most hazardous substance known to man" [2]. As the position statement adapted by the Health Physics Society explains, "Plutonium's demonisation is an example of how the public has been misled about radiation's environmental and health threats generally, and in cases like plutonium, how it has developed a warped 'risk perception' that does not reflect reality" [3].

As a result of this risk perception and ongoing debate surrounding environmental plutonium contamination, remedial action criteria are difficult to establish. By examining the data available before and after remedial actions taken at the three sites discussed in our report, we hope to present data that will illustrate that plutonium measured as aged deposition (older than several months) does not present as high a dose or risk as many had expected. The authors show that even though dose to the lung from inhalation (the primary pathway for the high-fired plutonium oxide particles present at these sites) is reduced, such a reduction is achieved at significant cost. The cost comes from damage to the environment, large expenditures per hectare rehabilitated, and the risk to occupational workers.

This paper specifically examines sites that are similar in many ways. These sites were chosen for their similarities to make comparisons. The sites are all desert in nature i.e., have low rainfall (all receive about 20 cm per year), have minimal vegetative ground cover, and have high summer temperatures. These sites are Palomares, Spain; the Nevada Test Site (NTS); and the Maralinga site in Australia. One significant difference, however, is that the Palomares site has been used continuously for residential and agriculture purposes since the plutonium remediation was completed. Maralinga is being remediated with the objective of returning the land to its former owners, but it will have some use restrictions for the remaining contaminated areas. Any decision to return the land being remediated by the United States Department of Energy (USDOE) at its Nevada sites, for public use, is in the distant future.

The plutonium source terms for each site are similar in that they were distributed as a result of the high explosive (HE) (i.e., chemical explosives) being detonated either purposefully or by accident. The resulting plume of vaporised plutonium was distributed in a cigar-shaped pattern dictated by the prevailing winds at the time of detonation. From this origin, the source material has the common characteristics of being created through a high-temperature process and rapidly oxidising to a very stable chemical state.

This high-fired Pu oxide is highly insoluble in the gastrointestinal system [4] and passes rapidly through the body when ingested. This accounts for the very small dose conversion factors for ingestion published by standard setting organisations such as the International Commission on Radiological Protection (ICRP) [5]. As a result of the nuclear nature of Pu-239, it decays primarily with a 5.15 MeV alpha emission, which means that a Pu-239 atom must be in close proximity to a cell before it can deliver any energy to that cell and potentially do harm.

This last finding is another reason why the ingestion pathway is of very minor concern. The epithelial cells lining the gastrointestinal (GI) system are replaced fairly rapidly. That fact coupled with the short residence time of the plutonium atoms in the GI system results in a dose that is very small and negligible. However, the situation is different in the lung, where small plutonium particles (e.g., $< 5\ \mu m$) can have very long residence times [4], and can also be in close contact with the lung's alveoli cells. This fact makes the lung the organ of concern when exposure to occupants of plutonium-contaminated land or re-occupants of Pu-remediated land are considered.

The route of exposure to the lung is through the inhalation process and an understanding of the factors and parameters that influence this pathway is necessary to predict the dose that an occupant of the contaminated land may receive. The primary parameters to be understood are source magnitude, particle size, and the combination of area size, wind speed, vegetative cover, and soil characteristics that give rise to the resuspension potential of the Pu-239 particles. Resuspension is largely an area phenomenon that requires a fairly large area affected by the contamination of interest, that is, an area where the contamination is on or very near the soil surface. Given these conditions when the wind blows, the particles can be picked up and consequently form an air stream of significant plutonium contamination. Several workers have discussed these dynamics and they have shown that receptors down wind will only be influenced by surface material within a certain distance. Shinn and Gouveia [35] in their report, "The Footprint Area Influencing a High Volume Air Sampler," have clearly demonstrated how the footprint fetch impacts a potential receptor. The worst case situation discussed in this report is bare soil, indicating that for a sampler at 1.13 meters high, 90% of the representative flux can be influenced by particles coming from as far away as 175 meters. When soil cover is present, the distances are reduced. Because of this area influence, measuring or determining Pu soil activity as an area average, i.e., Bq/m^2, is an important consideration.

The relationship of the above parameters is clearly discussed in many published reports, but we have chosen to follow the dose calculation method presented in the National Council on Radiological Protection and Measurements report (NCRP) 129 [6]. The model for lung inhalation dose used in that NCRP report is presented in Tables 1 and 2 along with the definition of the various parameters.

2. Methods

This paper focuses on a comparison of what has been learned from the measurements taken by various workers at these three desert sites to evaluate the resuspension parameters and how the remedial measures have affected them. We have examined the data available for each site and have compiled the data collected where available both before and after the remedial action to evaluate the effectiveness of the action. In the case of the Palomares site, which involved an accident, the data collected immediately after the contaminating event are very instructive.

Variables, which can be site and exposure scenario specific, such as the amount of air inhaled (m^3), along with the dose conversion factor (Sv/Bq), have been held constant for each of the before and after comparisons. We have used 8400 m^3 as the annual volume inhaled for adults living outdoors. We chose this number because the Maralinga Aborigines living a semitraditional life style live outdoors. The only change in the use of this parameter comes from calculating the dose for the lifestyle of hunting and hiking permitted to the returning Aboriginals. The volume inhaled has been adjusted to reflect the time individuals might spend in a worst case situation, which in this case would occur to individuals riding in the back of a following pickup truck while they were hunting kangaroo. As implied above in the case of the NTS and the Maralinga site, the calculated doses are hypothetical, as though people had been exposed or will be exposed at the levels indicated. However, this is not the case at Palomares. There, the values used and discussed have been experienced and continue to be experienced by the actual people who have been monitored (urine sampling and whole-body (i.e., lung) counting) for many years.

TABLE 1. Comparative dose assessment at the Maralinga and Palomares desert sites before and after soil remediation

	A	B	C	D	E	F	G	H	I
1	PARAMETER	Maralinga, Au before cleanup (Central Taranaki) and (Site FN)		Maralinga, Au after cleanup (Site FN) and (West Plume hunting scenario w/restrictions)		Palomares, Sp before cleanup	Palomares, Spain after cleanup (loc 2-2, 15y ave)	(loc 2-1, 4y ave) (9,10,31)	(loc 2-1, 1993)
2	**S**--Activity in Soil (Bq/g)	21 (24)	4.2 (34)	1.3 (25)	0.74 (23)	67 (10)	2.06	0.44	0.44
3	**E_r**-- (ratio between activity in respirable fraction of resuspended soil and activity in surface soil)	20			6				0.28 (31)
4	Arf -- Respirable fract. of resuspended soil- (Bq/g) **[D--soil deposition Bq/m^2]**	420	**[5.0 E+5] (34)**	**[2.0 E+4]**	4.44	**[1.2 E+6]**		**[5.0 E+5]**	0.123 **[3.3 E+4]** (31)
5	M--(mean dust loading in air (g/m^3) **[S_f-resuspension factor- m^{-1}]**	2.0 E-5 (34)	1.0 E-5 **[6.0 E-11]**	2.2 E-5 **[4.7 E-9] (windy conditions)**	0.08	**[1.0 E-7t+init] [1.0 E-9t+2m] [1.0 E-10t+yrs]**		**[2.6 E-10]**	7.0 E-5 **[1.0 E-10]** (31)
6	**C_{air}** -- Activity in Air (Bq/m^3)	8.4 E-3	3.0 E-5	9.1 E-5	3.6 E-3	1.2 E-1	5.36 E-5	5.6 E-5-	8.6 E-6 (31)
7	**R** -- (inhalation rate [m^3/y]) (26)	8.4 E+3	8.4 E+3	8.4 E+3	672.0**	8.4 E+3	8.4 E+3	8.4 E+3	8.4 E+3
8	AIA--Annual Intake of activity (Bq/y)	70.6	0.25	0.76	2.4	1008 init. 10.1 t+2m 1.0 t+yrs	0.45	0.47	0.072
9	**Df_{inh}**--[effective dose per unit intake (Sv/Bq)]	5.7 E-5*	5.7 E-5	5.7 E-5	5.7 E-5	5.7 E-5	5.7 E-5	5.7 E-5	5.7 E-5
10	equals **E_{inh}**--[effective dose from 1 yr of intake] (S v y^{-1})	4.0 E-3	0.014 E-3	0.043 E-3	0.14 E-3	57.5 E-3 init. 0.57 E-3 t+2m 0.057E-3 t+yrs	0.026 E-3	0.028 E-3	0.0041 E-3
11	**E_{inh}** in mSv/y	4	0.014	0.043	0.14	57.5 init. 0.57 t+2m 0.057 t+yrs	0.026	0.028	0.004
12	Dose Model--$E_{inh} = Df_{inh} \times C_{air} \times R$; C_{air} can be measured directly or estimated by $C_{air} = E_f \times S \times M$ or by the Resuspension Factor method where $C_{air} = S_f \times D$ (6) * The dose conversion factor (Df_{inh}) is based on NRPB experimental work on Maralinga Pu particals; NRPB recommended that 75% Y & 25 % W of the ICRP values be used (26). This value of 5.7 E-5 Sv/Bq has been used for all calculations for comparison reasons. ** Exposure while hunting (27)								

TABLE 2. Comparative dose assessment at the Nevada Test Site before and after remediation

	A	B	C	D	E	F
1	PARAMETER	Nevada Test Site, USA Double Tracks before and	after cleanup	Nevada Test Site, USA Clean Slate I before and	after cleanup	Reference
2	S--Activity in Soil (Bq/g)	4.52 Wtd. Ave. (1)	1.40 Wtd. Ave. (1)	4.81 Wtd. Ave. (1)	0.57 Wtd. Ave. (1)	(1) Steve Riedhauser, BN 29 Apr 99 (ref. 36)
3	E_f-- (ratio between activity in respirable fraction of resuspended soil and activity in surface soil)	0.69 (3)	0.69 (3)	0.69 (3)	0.69 (3)	(2) Desert Research Institute Air Sampling Report for Clean Slate I, 1998 (ref. 37)
4	Respirable fract. of resuspended soil-(Bq/g) **[D--soil deposition Bq/m^2]**	**[6.8 E+4 Wtd. Ave. (1)]**	**[2.1 E+4 Wtd. Ave. (1)]**	**[7.2 E+4 Wtd. Ave. (1)]**	**[8.6 E+3 Wtd. Ave. (1)]**	(3) Shinn-1997 Resuspension at the Tonapah Range (ref. 29)
5	**M**--(mean dust loading in air (g/m^3) **[S_f-resuspension factor- m^{-1}]**	5.9 E-6 (4) **[1.3 E-10 (3)]**	8.2 E-6 during (4) 4.8 E-6 after (4) **[1.3 E-10 (3)]**	3.5 E-6 (2) **[1.3 E-10 (3)]**	7.1 E-6 **[1.3 E-10 (3)]**	(4) Desert Research Institute Air Sampling Report for Double Tracks. 1997 (ref. 33)
6	C_{air} -- Activity in Air (Bq/m^3)	1.1 E-7 (5) measured **[8,8 E-6 calc. from S_f & D]**	2.4 E-5 meas during (4) 1.11 E-7 meas. after (5) [2.7 E-6 calc fm S_f & D]	7.2 E-6 meas. (2) [9.4 E-6 calc fm S & D]	1.6 E-4 meas. during (2) 2.8 E-6 meas. after (2) **[1.11 E-6 calc after]**	(5) DOE/NV Double Tracks Closure Report-1997 (ref. 32)
7	**R** -- (inhalation rate (m^3/y)) (26)	8400	8400	8400	8400	Inhalation rate for standard man living outdoors 365 days per year (26)
8	AIA--Annual Intake of activity (Bq/y)	9.2 E-4 fm meas. **[7.4 E-2 fm S_f& D]**	2.0 E-1 during (4) 9.3 E-4 after (5) [2.3 E-2 calc fm S & D]	4.03 E-2 [7.9 E-2 calc. fmS & D]	1.34 during excav. (2) 2.3 E-2 after excav. (2) [9.3 E-3 calc after, S& D]	Note: At Double Tracks and Clean Slate I, DRI collected air samples and dust samples at the edges of the area undergoing excavation in the direction of the prevailing winds. Sampling was done months before, during and after the excavation period. Water was used to supress dust during excavation and a fixative was applaid after. These methods/agents were fairly effective as the observed E_{inh} is very small. The during values being the only ones showing any modest resuspension.
9	**Df_{inh}**--[effective dose per unit intake (Sv/Bq)]	5.75 E-5 *	5.75 E-5 *	5.75 E-5 *	5.75 E-5 *	
10	**E_{inh}**--[effective dose from 1 yr of intake] (S v y^{-1})	5.2 E-8 calc fm meas. **[4.2 E-6 calc.]**	1.1 E-5 during excav. 5.3 E-8 after excav. [1.3 E-6 calc fm S & D]	2.3 E-6 before [4.5 E-6 before, S & D]	7.6 E-5 dur excav 1.3 E-6 after excav [5.3 E-7 after, S&D]	
11	**E_{inh} in mSv/y**	5.2 E-5 [4.2 E-3 calc.]	1.1 E-2 during excav. 5.3 E-5 after excav. [1.3 E-3 calc fm S & D]	2.3 E-3 before [4.5 E-3 before, S & D]	7.6 E-2 dur excav 1.3 E-3 after excav [5.3 E-4 after, S&D]	
12	Dose Model--E_{inh} = Df_{inh} X C_{air} X R; C_{air} can be measured directly or estimated by C_{air} = E_f X S X M or by the Resuspension Factor method where C_{air} = S_f X D (6) * The dose conversion factor (Df_{inh}) is based on NRPB experimental work on Maralinga Pu particals; NRPB recommended that 75% Y & 25 % W of the ICRP values be used (26). This value of 5.7 E-5 Sv/Bq has been used for all calculations for comparison reasons.					

3. The Three Sites

3.1. PALOMARES, SPAIN

On January 17, 1966, a U.S. Air Force B-52 bomber collided with its tanker and exploded above the town of Palomares, Spain. Of the four nuclear weapons onboard the bomber, three impacted very near the town and the fourth fell into the sea. The chemical high explosives of two of the four weapons detonated on impact and bracketed the residential area. Plutonium particulate contamination was distributed in varying degrees over a 226 ha area (~560 acres) consisting of brush land, farmland, and an urban area. The other two weapons were recovered intact.

A set of tiered criteria applied to the cleanup (Table 3) was completed by May 1966. In the most heavily contaminated area, crops and soil were removed to a depth of approximately 10 cm. These materials were then packaged and shipped to the United States for disposal. The next level of contamination required that canes be burned on the beach, crops buried, and soil plowed to 30 cm. The remainder of the contamination required that soil be plowed to 30 cm where possible [7].

TABLE 3. Cleanup criteria for the three desert sites

Site	Principal Criterion	Comments
Palomares, Spain 1/17/66; //66 * (7)	1180 kBq/m^2 -soil removed to 10 cm 118 kBq/m^2 -soil plowed to 30 cm 11.8 kBq/m^2 -some soil plowed to 30 cm	Approximately 1000 m^3 of soil was placed in 4810 barrels and shipped to the USA. Crops were removed and burned or buried on the beach 226 ha remediated
Maralinga, Australia (British Test Site) 9/56-4/63; //67//96-99 (7)	part./frag.--remove if >100 Kbq; if [illegible] 20kBq averaged Not More Than 1/10 m^2; TRU in Soil (1ha ave.) / Am-241/m^2 Taranaki - 1.2 Bq/g / 3 kBq/m^2 TM-100 - 2.2 Bq/g / 1.8 kBq/m^2 TM-101 - 1.7 Bq/g / 4.0 kBq/m^2 Wewak - 0.7 Bq/g / 1.8 kBq/m^2	activity of the part./frag. are for Am-241; TRU is primarily Pu-239,240; The guideline varies primarily because of the difference in the Pu/Am ratio at the specific sites. 147.4 ha remediated

Nevada Test Site
Double Tracks Event
5/63; // summer 96
(17)
	DOE Order 5400.5, Ch.4
	1 mSv/yr (100 mrem/yr)
	Remove Soil >7.4 Bq/g (200 pCi/g)
	Pu-239,240; ave. over 1ha
		Remove all fragments
		2.6 ha remediated

*Operational period or event date; **//69 Year or period of time cleanup took place. Some locations had more than one cleanup.

Upon completion of the cleanup and soil restoration of the farmland, a research/monitoring programme began. For reference, the contaminated area has been subdivided into three zones (zones 2, 3, and 5), corresponding to the number assigned to the weapons and where they impacted. Zone 5 is the urban zone (Fig. 1). This programme consisted of air sampling, soil sampling, crop sampling, urine sampling, and lung counting of residents. Members of the Spanish Junta De Energia Nuclear [8, 9, 10] carried out this work.

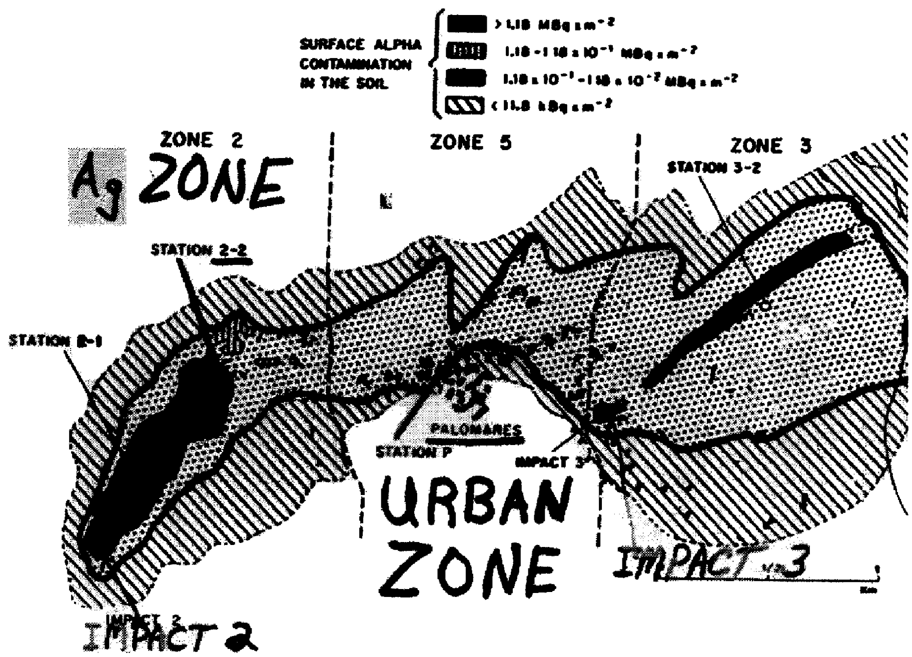

Figure 1. Palomares area with original surface contamination levels and location of air sampling stations. Black spots show the location of houses.

On the day of the accident 485 people were present in Palomares. However, because it was a holiday, few were working in the nearby fields. Over the 22 years of monitoring by the Spanish Government, 229 additional people have been included, bringing the total to 714. They either moved into the area or were born after the accident. Of this number 590 had urine sample results lower than the minimum detectable activity (MDA) of 0.37 mBq/d and 124 had values equal to or higher than the MDA. Of the 124, 29 showed sample contamination, because samples were collected in Palomares. Beginning in 1967, residents travelled to Madrid for sampling and annual examination. Estimations of dose were performed on those 55 people who were considered to really have suffered internal contamination. Lung counting, using the most sophisticated detectors for the time, indicated no plutonium level above the MDA of 814 Bq. From the urine sampling, it was determined that 45 of the people had experienced acute inhalation of particles. Chronic inhalation at the average concentration measured in air could not support the quantity observed in the urine. From this the date of intake could be assigned. For the remaining 10, who were not in the area on the day of the accident, the date of intake considered to be the most likely was assigned. The 70-year whole-body dose committed effective dose equivalent (CEDE) for the initial 45 residents ranged from 20 to 200 mSv (0.29-2.9mSv/y). On the basis of the suppositions for the group of 10, a range of from 35 to 180 mSv is estimated. The remaining population (659 residents) is estimated to have received less than 20 mSv, CEDE. There were 10 residents who were 15 years or younger. Of these only one (<1 yr on Jan. 17, 1966) has a CEDE higher than 200 mSv (242 mSv); the other 9 are in the range of from 49 to 157 mSv [11].

Aerosol measurements [8, 9, 10] of Pu-239 and Pu-240 (Table 4 and Figure 2) exceeded the detection limit (1.8 E-6 Bq/m^3) for all years at all sampling stations, except in the urban zone for the years 1971, 1972, 1975, and 1976. The frequency of air samples exceeding the detection limit diminished with time until a hilly and uncultivated parcel was plowed and transformed into a cultivable parcel in 1974. Additional cultivation in the early 1980s also contributed to increasing air concentration [31]. At station 2-2, near the newly cultivated land, higher annual concentration averages have occurred throughout the period. The maximum concentration occurred in 1967. For the urban area, the average concentration has been below a hundredth of the derived air concentration (DAC) (calculated for the public) for Class Y Pu compounds. In the farming zone (2-2), the average concentration was below one-tenth of the DAC (public) for Class Y Pu compounds. For the period 1966-69, the average concentration was below the DAC. To be on the conservative side, doses calculated for various organs were based on an average mass activity diameter (AMAD) of 1.0 μm for the particles inhaled. The bone surfaces received the highest potential committed dose equivalent; the sum for each of the 15 years (1966-80) has a value of 0.56 mSv for the urban zone and 5.42 mSv in zone 2-2. In the five organs of interest, the contribution of the committed dose equivalents to the potential CEDE during the 15-y period is 0.054 mSv in the urban area and 0.52 mSv in zone 2-2 [9].

The Palomares region of Spain is typical of Mediterranean agricultural areas that receive marginal rainfall (~20 cm/y) and require irrigation to sustain crops. Common crops of the area are tomatoes, grain, and alfalfa. The plutonium concentration observed for washed tomatoes is 0.15 Bq/kg and is generally a factor of 30 to 40 higher for the plants, stalks, etc. of tomato plants, barley, and alfalfa. The soil-crop concentration ratios are of the order of 10^{-4} for tomatoes and 10^{-3} for the plants and the components of barley and alfalfa. The annual CEDE to individuals ingesting tomatoes is 1.5 μSv. Other pathways, i.e., alfalfa, meat, or milk, to humans are even less [12].

The data and experiences recorded for Palomares are very important. To date the people of Palomares belong to a very well-documented group, that since 1966 have lived and worked continually as farmers in a plutonium-contaminated environment.

TABLE 4. Composite Data Table, Palomares, Spain 1966-88

Pu-239 + 240 in Samples or Dose from Pu 239 + 240 in Organs*						
Parameter Reported	Urban Zone		2-2 Zone (Agr.)		2-1 Zone (Agr.)	
Soil surface contam. MBq/m2	0.012		0.012 to 1.2		0.012 to 1.2	
Concentration in Soil (Bq/g Ave. Max. Min. (28)	0.13 0.30 0.02		2.06 3.31 0.80		0.44 1.60 0.03	
Sample Year	Surface Soil Conc. MBq/m^2	Airborne ☞Bq/m^3	Surface Soil Conc. MBq/m^2	Airborne ☞Bq/m^3	Surface Soil Conc. MBq/m^2	Airborne ☞Bq/m^3
1966	14.8		44.8			
67	4.1		441.8			41.8
68	2.6		21.8			15.2
69	0.0009	2.6	0.19	142.0		7.0
70	5.9		2.2			161.0
71	< 1.8		2.2			
72	0.013	< 1.8	0.27	10.4	4 year average = 56.0 μBq/m^3	
73	0.0004	2.2	0.01	3.0		
74	4.1		8.1			
75	0.0006	< 1.8	0.06	16.3		
76	0.002	< 1.8	0.01	4.4		
77	0.001	5.6	0.01	11.8		
78	2.2		16.7			
79	0.003	5.6	0.02	19.2		
80	28.1		32.9			
81	0.004	14.3	0.30	46.6		
(27,28) 83	0.004	9.1	0.31	87.9		
Shinn** * 1993					0.033	8.5
Potential CEDE for 1st 15 years from chronic inhalation (1966-80) Five Organ Total** (mSv)	(lung-0.023) 0.054 mSv		(lung-0.22) 0.522 mSv		(lung-0.004 mSv/y Shinn, 1993)	
Percent of 1 mSv/y Effective Dose Equivalent (26,27)	5.4 %		52.3 %			
(SE70) 70 y CEDE from the Acute Inhalation Exposure at the time of the accident for 45 residents (29)	20-200 mSv (This data doesn=t apply to zones)				***Data from Shinn who independently completed a sampling trip in Zone 2-1 during 1993	

*Data from Iranzo, 1987 & 1994
Soil Surface Samples were collected using a square metal grid 25 cm x 25 cm x 5 cm in depth.
Soil samples were collected from study plots 50m X 50m designated for repeat sampling.
Aerosol sampling using high-volume samplers (1 m^3/min) were at various locations.
** Five Organs consist of: Lung, Bone Surface, Liver, Red Marrow, & gonads.

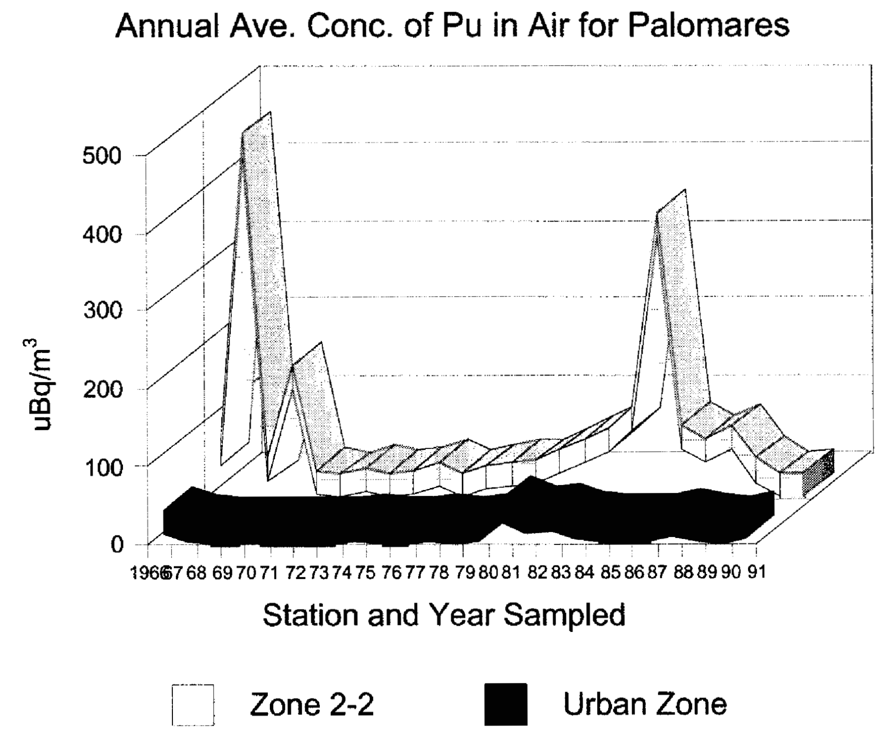

Figure 2.

3.2. NEVADA TEST SITE

The Nevada Test Site and environs have approximately 8500 acres contaminated with plutonium >3.7 Bq/g on the land surface because of one-point-safety tests and plutonium-dispersion tests [13]. During the summer of 1996, an interim cleanup action was completed at the Double Tracks event site. An additional site (Clean Slate 1) was cleaned up in the spring of 1997. Both were part of the Operation Roller Coaster series. These two sites conformed to the criteria outlined in Table 2. However, cleanup activities are considered by officials to be interim actions because no definitive plutonium-in-soil standard or guide exists. USDOE Nevada (NV) has recently negotiated a Federal Facility Agreement and Compliance Order (FFACO) with the State of Nevada. There has not been time for the State (lacking a federal plutonium-in-soil standard) to decide on a Nevada standard for USDOE to use. It has been contemplated, as suggested by the Double Tracks Environmental Assessment [14], that the cleanup ~~on~~ criterion, expressed as a concentration, is likely to be 14.8 Bq/g, but could be as low as 3.7 Bq/g. The value of 7.4 Bq/g as used is expected to be conservative [15].

The primary dose limit specified in USDOE Order 5400.5, Chapter 4, for all USDOE activities (including remedial actions), is expressed as a CEDE. This specification comes from the ICRP 26, 30 risk-based system, which requires a summation of doses to various organs of the body using weighting factors to be applied to each major tissue and organ. Exposure (as a consequence of USDOE activities) to members of the public from all radiation sources is not to exceed 1mSv/y. Though USDOE and others have established guidelines for thorium and radium in soil, guidelines for residual concentrations for other radionuclides in soil have to be derived from the basic dose limit. Creating an environmental pathway analysis, in which site data and default parameters are fed into a dose prediction model, does this. In USDOE's case they are required to use the RESRAD computer model developed by Argonne National Laboratory [16].

Because the "correct" land use and resultant pathway selection can be in the "eye-of-the beholder," this method generally causes considerable discussion among regulators, stakeholders, and management. This is happening with the Nevada State Division of Environmental Protection (NSDEP). The USDOE Nevada office had chosen to use a rancher's land-use scenario and resultant pathway assumptions [17], but the director of NSDEP has made it clear that he is not readily accepting the USDOE/NV land-use scenario [18]. He has implied that future negotiations must be held before the matter of the "correct" land-use and resultant cleanup criteria can be established.

3.3. MARALINGA, AUSTRALIA

Maralinga was one of three sites used by the British for nuclear weapons testing in Australia [19]. The Royal Commission Hearings in the mid-1980s initiated the events leading to the current remedial programme. A Technical Advisory Group (TAG) was formed upon a recommendation at the hearings to investigate the aspects of future remedial action and to recommend cleanup options for the Australian government to consider. The TAG report offered a group of options. From these, the government selected option 6c, which in brief, suggested surface-soil excavation and burial in deep trenches on site. Pits located a distance from Taranaki have been exhumed. Of the 21 shallow burial pits filled with plutonium-contaminated metal debris, 11 were treated by *in situ* vitrification; 10 were exhumed and their contents deeply buried. The posting of signs to limit activities in the large, unremediated downwind fallout plumes has been accomplished [20].

This project will cost about $A 104 million and will remove over 322 000 m^3 of soil [21]. The project's objective is to return use of government-controlled lands to former native owners. The soil-removal criterion for rehabilitation was based on the annual risk of fatal cancer associated with the inhabitants inhalation or ingestion of contaminated soil. This risk is not to exceed one in ten thousand accumulated by the fiftieth year. It was considered that the soil contamination contour corresponding to an annual committed dose of 5.0 mSv is the borderline between acceptable and unacceptable risk [20].

Large amounts of metal and other materials were immediately adjacent to the explosive device of each of the 12 one-point safe trials conducted at Taranaki. Because of these materials, an area of about 2.5 km^2 was extensively contaminated with fragments and particles (the difference being visible or not visible to the naked eye). Concern that the fragments might cause or enter a wound on the bare feet of the inhabitants largely dictated the extent of the area for soil removal. The remediation plan not only specified the various earth works, but it also had to deal with restrictions on land use. For example, in the plume areas where excavating such large areas was not feasible, only casual travel and hunting will be allowed. The option for the Aboriginal people to live in these areas was relinquished because of the cost and extensive environmental damage required to reduce the average plutonium concentration to an acceptable risk level for a semitraditional Aboriginal lifestyle. The Aboriginal landowners, who were able to participate in the development of cleanup objectives and plans, readily agreed to the hunting and travel restrictions. Their participation helped to prevent severe and extensive damage to the environment.

4. Results

Table 3 and Figure 2 display the monitoring data gathered for the Palomares site by Iranzo *et al.* [9,10]. These data are particularly compelling because, as has been mentioned, they are for a population that was not only present at the time of the incident, but one that has continued to live (now 33 plus years) in the contaminated area since the event. As was discussed above, only 45 individuals of those who were out of doors in the near vicinity of the HE explosive dispersal have had "measurable" plutonium in their urine. The remaining 440 have never had sufficient quantities in their excreted urine to detect plutonium. Over the years, these individuals, along with an additional 229 plus the miscellaneous exposed 10, add to 714 people who have lived and worked in and around the contaminated area illustrated in Fig. 1. The monitoring data displayed show low levels of airborne contamination with some years spiking because of increased disturbance of the nearby soil because of pond construction and the cultivation of new ground. However, even with these activities, the airborne concentration of plutonium remains low and the calculated dose, calculated because it is so low it cannot be directly measured via bioassay, is only a fraction of the dose published as acceptable and safe by international organisations [5, 22]. It is instructive to examine the data in Table 1, row 5, column F, which displays the resuspension factors measured by Iranzo *et al.* [10] at the time of the fresh deposition and then as it aged at 2 months and years. It is also observed in cell F-11 that the corresponding E_{inh} (effective dose for inhalation) also drops dramatically.

At Maralinga the remediation, which is nearly complete, was driven by criteria developed for a returning Aboriginal community that would be living a semi-traditional lifestyle. These people live in a camping environment that creates considerable dust. The few dust loadings that were measured were high. The dust loading used in the dose-projection calculations was based on limited measurements indicating a value of 1.0 mg/m^3 for adults. A dust loading this large indicates a very dusty environment and is generally associated with major dust storms or severe dust-raising activities. The authors of NCRP report 129 present data from various workers who found annual averages of 28 and 75 ☞g/m^3 for undisturbed conditions for rural and urban locations, respectively. The report also shows data collected from behind a tractor, which had a median of 15 mg/m^3 and a range of from 0.3 to 200 mg/m^3 (mass loading values over 0.15 mg/m^3 are considered nuisance dust) [6]. The annual effective dose E_{inh} calculations for the semitraditional Aboriginal lifestyle used 1 mg/m^3 dust-loading and enhancement factors as indicated in Table 1. The enhancement factor (E_f) is defined as the ratio of the activity concentration of the respirable fraction to the activity concentration of the sample as the whole. The E_f was determined for soil samples from separations performed with a Bahco microparticle classifier. The E_f for the resuspended material was determined from cascade impactor data. It is important to note that the "enhancement factor" as defined earlier contains no depth-related information. It merely relates the activity concentration in the inhalable fraction of a given soil sample to the activity concentration of the bulk sample, whether the sample is of surface soil or a suspension in air.

To convert from activity per unit surface area to activity per unit mass, it is necessary to know the profile of activity with depth, as well as the soil density. At Maralinga, measurements show that in undisturbed areas, most of the activity is in the top 10 mm of most samples and at least 85% is in the top 20 mm. Average soil density was found to be 1.7 g/cm^3. From these data and for the purposes of calculation, it is assumed that, in undisturbed areas, all the activity is in the top 10 mm of soil and that it is uniformly mixed throughout that depth. On the basis of these assumptions, an airborne dust concentration of 1 mg/m^3 corresponds to a resuspension factor of 6×10^{-8} per m for an enhancement factor of unity.

Using the model parameters presented in Table 1, Williams *et al.* [23] calculated 273 mSv/y for central Taranaki using parameters specific to a camping lifestyle (i.e., large mass loadings). Johnson *et al.* [24] have published a dose commitment rate indicated in Table 1, in cell B-11 of 4 mSv/y for adults for central Taranaki. The difference can be attributed to undisturbed soil conditions at the time of the measurements as indicated by the low mass loading, 0.2 $\mu g/m^3$ (Table 1; cell B-5). The resuspension factor (S_f) determined by Johnson for a nearby area (site FN- Fig. 3) within central Taranaki before cleanup (cell C-5) is also very low. Soon after cleanup of this area, approximately 10 years later, a similar determination of S_f was made by Shinn (cell D-5) [25]. The value in Table 1 is for windy conditions; the value for normal conditions is an order of magnitude lower (3.2 E-10 m^{-1}). Table 1, (cell D-5) also indicates a mass loading in the $\mu g/m^3$ range under windy conditions and very soon after excavation of the area was completed (i.e., under disturbed conditions).

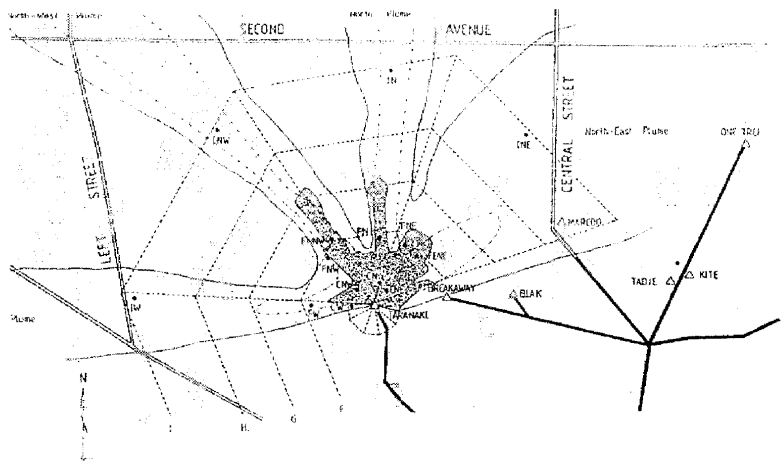

Figure 3. Map of Taranaki Area showing location of FN

Measurements and dose estimates have also been made in the downwind plumes using the model and aboriginal life style discussed above. The results of the calculation have been published by Heywood *et al.*, [26]. The input parameters were similar to those of Williams [23], e.g., mass loadings of 1 mg/m^3 for the adult and large enhancement factors, i.e., greater than unity. The value calculated is 3.0 mSv/y for the adult in the northwest plume; for the 10-year-old child it is about 20% more. In Table 1, column E, we have shown the results, using the same method as previously described, for the "hunting scenario."

The Aboriginal stakeholders have agreed to restrictions that limit their use of the land to walking and hunting, but no camping. The occupancy factor for this scenario has been adjusted according to the time spent hunting as determined by Palmer and Brady [27].

The USA has conducted tests at Nevada that were similar to the Vixen trials at Maralinga, and the USDOE has recently engaged in remedial actions at two of these event sites, Double Tracks and Clean Slate I of the Roller Coaster Series. The series consisted of four events, jointly sponsored by the US and UK, at the Tonapah Test Range (TTR), near Tonapah, NV. These tests were conducted to study the safety aspects of the transportation and storage of nuclear weapons. The data displayed in Table 2, attempt to show the changes in resuspension and the resulting E_{inh} before and after the remedial actions. Like Maralinga, and unlike Palomares, the sites have not been used for many years. Prior to remediation the surface plutonium particles are in a stable and undisturbed desert shrub environment. As the data suggests, the various parameters indicating the ability to resuspend, i.e. S_f and M were very low prior to the commencement of any earth works. Because the USDOE used water spray periodically to suppress dust during the excavation [32] and a soil fixative upon completion, the mass loading, M and S_f remained low. Observable plutonium was measured on air samples placed at the perimeter on three sides covering the majority of the wind regimes recorded. However, the E_{inh} would not exceed 10% of the annual effective dose to the public, if they stood at the excavation boundary continually for a year. The scenario is impossible because the excavation and shipping only lasted for less than a two-month period, after which the indicated E_{inh} dropped by a factor of 58.

Resuspension studies conducted year around by Shinn *et al.*, [29] at TTR show that weathering, e.g., the spring freeze/thaw cycle, etc., does create variability in the S_f. The range is over an order of magnitude with the median at 1.3 E-10 m^{-1}. All the S_f values measured at the three sites reviewed in this paper mostly fall within this range (1.0 E-9 to 1.0 E-10 m^{-1}). The exceptions were the initial observed S_f at Palomares (E-7 m^{-1}) and the S_f measured at central Taranaki before cleanup (E-11 m^{-1}). Shinn and others [30, 6] have also reported that this range is generally observed at many other sites around the world, e.g., at the Marshall Islands, at Chernobyl, and at other locations on the Nevada Test Site.

5. Conclusions

- Investigations of all three sites studied in this report show that within a short time period, e.g., a few months, plutonium-contaminated debris becomes fairly stable, even when the soil surface is periodically disturbed, as in the case of farming at Palomares, and of excavating at Maralinga and TTR. This finding is important in considering cleanup criteria, because it suggests that even after severe disturbances associated with remedial actions and or farming, the soil surface soon returns to a condition that limits resuspension of plutonium particles.
- From the experience and data observed at the three sites, it is possible to estimate conservatively the deposition required to deliver 1 mSv/y (annual dose commitment to a member of the public) to potential inhabitants who spend their entire year outside. The following are the required conditions: The Df_{inh} is for very insoluble particles, such as high-fired Pu oxide; the S_f falls within the range observed for plutonium material over several months old; and the mass loading is not something very abnormal (≥ 1 mg/m^3). With these general conditions met, one should be able to dependably solve for D (Soil Deposition, Bq/m^2). The solution indicates that it would require the soil deposition in the low MBq/m^2 range to come close to the dose limit for a member of the public.
- As Palomares is the only site where members of the public routinely live and work in a plutonium-contaminated environment, we learn that indeed working and living in these environments yield only fractions of the annual public effective dose. The data that Shinn [25] collected in 1993 in an agriculture area indicate less than 1% of the annual dose to a member of the public, and this is if they were at that location 365 days a year. To date it has not been reported that members of the community inhale or ingest enough plutonium from the contaminated soil to be detected in bioassay samples.
- If the Palomares experience is valid, and the S_f levels observed around the world are reasonable indicators of potential exposure, then remedial actions looking at only a few Bq/g are very, very conservative (see Table 1).
- On the basis of the actual human experience observed at Palomares, Iranzo *et al.* [12] recommended that the International Atomic Energy Agency (IAEA)

consider 1 man Sv of collective effective dose equivalent as a guideline to exempt quantities for practical application. They further recommend that for the crop types experienced, 120 to 1200 kBq/m^2 would be an appropriate intervention level, depending on the size of the contaminated area. The larger the area, the higher the intervention level.

- It is also observed for Palomares that only those individuals who were outdoors and in the near down wind area at the time of the accident received doses approaching and exceeding the annual dose limit for the public and had measurable plutonium in their urine samples. Resuspension of the deposited Pu particulate gives minimal dose to the receptor's organs. The CEDE is much less than the specified limits. Working, living, and eating products grown in a contaminated area of the magnitude experienced at Palomares yield extremely small doses that are much less than the accepted standards.

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